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Annealing Performance of Cu_2ZnSnS_4 (CZTS) Thin Film Solar Cells Fabricated by Chemical Bath Deposition

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ABSTRACT

Cu_2ZnSnS_4 (CZTS) thin films have high optical absorptive, direct band gap and non- toxic earth abundant composition. (CZTS) thin films were prepared by chemical bath deposition (CBD).These films were annealed at ~680^0 and further characterized by X-ray diffraction (XRD), Scanning electron microscopy (SEM), and UV-Vis measurements. X-ray diffraction (XRD) studies shows that the grain size and structural information of CZTS films. Scanning electron microscopy (SEM) studies shows that the formation of densely packed, compact and surface defects of CZTS films. The optical measurements will show the band gap of these films is around 1.4 ev – 1.6 ev, and then its optical absorption coefficient is on the order of above 10^4 cm^{-1}.A type of solar cell structure is glass/Mo/CZTS/CdS/i-ZnO/ZnO:Al/Al-grid are effectively fabricated. In this cell has a good agreement with short-circuit current density (I_{sc}), open-circuit voltage (V_{oc}),a fill factor(FF) and efficiency under AM 1.5(100mW/Cm^2) illumination. The CZTS thin films have been showing semiconductor behavior with p-type electrical conductivity. Further photovoltaic activity of these films was studied by after formation.

Keywords: CBD, Cu_2ZnSnS_4, semiconductor, open-circuit voltage.

1. INTRODUCTION

CZTS is one of the technologically important materials, which has been the subject of extensive studies because of its typical chemical, electrical and optical properties such as suitable band gap energy, excellent transparency and refractive index in the visible light and good semiconducting properties. Copper zinc tin sulphide has received an enormous attention because of its potential use in photovoltaic and gas sensing applications. The high efficiency and non-toxicity[1] of CZTS have stimulated intensive research on environmental applications. CZTS have high absorption coefficients (>104 cm^{-1}) in the visible range of the electromagnetic spectrum, and only a few micron thick films can absorb all the radiation above the band gap. CZTS thin films have been synthesized using co-evaporation[2], RF-sputtering[3], chemical bath deposition[4], electrodeposition[5], spray pyrolysis[6], pulsed-laser deposition[7], spin coating[8] from precursor solutions, and casting from nanocrystals dispersions. Recently, CZTS solar cells have been shown to achieve a power conversion efficiency of 10.1%.[9].

In this study, we studied CBD coated CZTS thin films by changing solvents. Precursor was fixed with all chloride forms with thiourea. Thiourea was added to supply sulfur content into the precursor. Finally photovoltaic activity of these films was studied by after formation.

2. EXPERIMENTAL DETAILS

CZTS thin film was deposited on precleaned glass substrate using CBD. The substrate was cleaned in acetone and deionized water. Finally it was dried at 90°C for one hour. The bath solution consists of required amount of copper chloride (2M), Zinc chloride (1.2M), tin chloride (1M) and thiourea (8M) were added in a mixture solvent ethanol/water(30:70).Clear yellow sol – gel was formed after being stirred at room temperature for ten minutes. The substrate was dipped in the solution for 24 hours. A thin coating with white Coloration appeared on the substrate. The coated substrate was removed at the end of deposition, washed in deionized water, dried in air at 110^0C temperature. Finally, the annealing temperature was elevated to 500^0C for growing polycrystalline CZTS thin films and they were subjected to morphological, optical and photovoltaic characterizations.

3. RESULTS AND DISCUSSION

3.1 XRD analysis

The structural analysis of CZTS thin films was carried out by using X-ray diffractometer. The X-ray diffraction patterns of the CZTS thin films, on glass substrates are shown in Figure 1.The XRD analysis shows that the thin films are kesterite phase CZTS with lattice parameters a = 5.409Ǻ and c = 10.642Ǻ which is almost in agreement with the standard data from JCPDS card No 26-0575. The planes are oriented in the direction (002), (101) and (314). The films exhibit tetragonal crystal structure. The highest intensity peak corresponds to (002) preferred orientation. The (002) peak is stronger than other peaks. In general, the preferential orientation of the films is along the (002) direction.

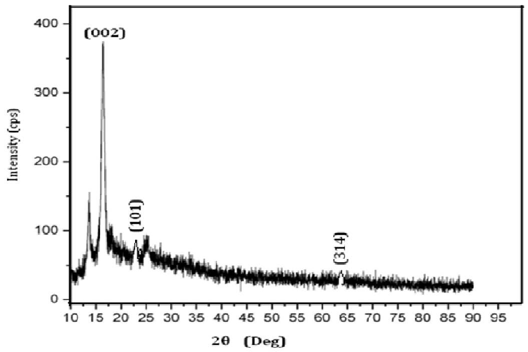

Fig.1 XRD pattern spectrum of the Cu_2ZnSnS_4 film

3.2 Morphological analysis

Fig.2(a-c) shows the surface (SEM) image of annealed CZTS film. Annealed CZTS film composed of particles having size ranging from 250 to 300 nm. Particle size and crystallite size are different. Crystallite size is always smaller than particle size because a particle constitutes many crystallites. Moreover, the efficiency of TFSC increases with increasing crystal size of the absorber layers[10].

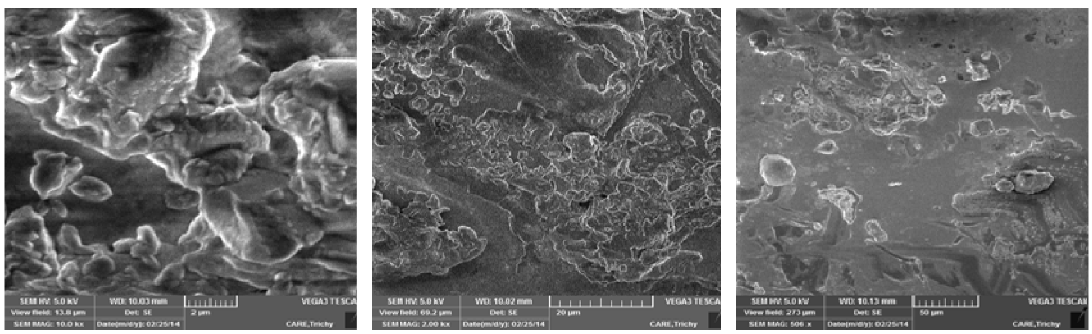

Figure 2: The SEM images of the CZTS thin film (a) 20 µm (b) 20 µm (c) 50 µm

3.3 Optical Properties

Optical band gap energy (Eg) of synthesized films was calculated and plotted. Eg was attained from UV-VIS analysis based on the following equation.

$$\alpha h\nu = (h\nu - Eg)^2 \quad (2)$$

Where, α is the light absorption coefficient, h is the Planck constant and ν is the frequency. Extrapolation method in conjunction with the above equation produces Eg The band gap is estimated to be 1.51 eV by extrapolating the straight line part of the $(\alpha h\nu)^2$ versus $h\nu$ curve to the intercept of the horizontal axis. This value is quite close to the theoretical optimal value for a single-junction solar cell[11].

4. CHARACTERIZATION OF THE CELL

CZTS solar cells with an active area of 0.46 cm2 were fabricated and characterized. All cell parameters were relatively low. The current density-voltage (*J-V*) characteristic of the best solar cell efficiency is 2.63% achieved.

5. CONCLUSIONS

CZTS thin film was successfully deposited by a CBD method. The annealed film showed large densely packed grains.

This film possesses a high optical absorption coefficient and optical band gap near to the ideal band gap of the absorber for single junction solar cells. The film could be a suitable photovoltaic material from the perspective of optical properties. Desired composition was achieved for the CZTS thin film after preannealing at low temperature. Loss of tin was observed after annealing at higher temperature, as should be solved for high-performance thin film solarcells. The best solar cell showed a low conversion efficiency of 2.63%. Optimization experiments are in progress and expected to lead to a significant improvement.

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